Interaction of 3–level atom with radiation

L.Accardi, K.Imafuku, S.V.Kozyrev

April 25, 2001

Abstract

The interaction of 3–level system with a quantum field in a non–equilibrium state is considered. We describe a class of states of the quantum field for wich a stationary state drives the system to inverse populated state. We find that the quotient of the population of the energy levels in the simplest case is described by the double Einstein formula which involves products of two Einstein emission/absorption relations. Emission and absorption of radiation by 3–level atom in non–equlibrium stationary state is described.

1 Introduction

In the present paper we consider a 3–level system (for example an atom) interacting with radiation. In the work [1] we apply the technique developed here to describe stimulated emission for 3–level atom interacting with radiation in a non–equilibrium state. In the work [2] a 2–level quantum system interacting with quantum field was considered. In [3] a stationary nonequilibrium state for an n–level system interacting with a nonequilibrium quantum field was obtained.

We show that for such a system application of the stochastic limit allows to obtain an interesting effect of inversion of population: for a special choice of the state of reservoir the system relaxes to stationary state where the population of the level of the system with higher energy will be larger than the population of the level with lower energy.

We obtain an equation for the number of photons emitted and absorbed by the system.

We investigate the examples of 2–level and 3–level systems. We show that for a 2–level atom the emission in the stationary regime equals to absorption. For a 3–level system emission and absorption of radiation are controlled by the state of the field. We find that for a 3–level system in a stationary nonequilibrium state two regimes are possible: the emission and the absorption regime. In the emission regime the total number of quanta in the system increases, and in the absorption regime it decreases. These regimes are controlled by the function $\beta(\omega)$ in (3). For example a 3–level system with energy levels $\varepsilon_1 < \varepsilon_2 < \varepsilon_3$ is in emission regime when

$$\beta(\varepsilon_2 - \varepsilon_1) + \beta(\varepsilon_3 - \varepsilon_2) > \beta(\varepsilon_3 - \varepsilon_1)$$

and is in absorption regime when the opposite inequality holds.

In the emission regime the 3–level system converts radiation with the frequency ω_2 into radiation with the frequencies ω_1 and ω_3 and vice versa in the absorption regime.

This means that this stationary state of the 3–level system gives an example of dissipative structure in the Prigogine sense [5].

For an equilibrium state of the field, i.e. when the function $\beta(\omega)$ is linear, the system is in equilibrium with radiation.

The interaction of a quantum system with a quantum field is described by an Hamiltonian

$$H = H_S + H_R + \lambda H_I \tag{1}$$

The system degrees of freedom are described by the system Hamiltonian H_S.

The radiation degrees of freedom are described by the Hamiltonian

$$H_R = \int \omega(k) a^*(k) a(k) dk \tag{2}$$

where $a(k)$ is a bosonic field with a Gaussian state of the following form

$$\langle a^*(k) a(k') \rangle = N(k)\delta(k-k') = \frac{1}{e^{\beta(\omega(k))} - 1}\delta(k-k') \tag{3}$$

and $\beta(\omega(k))$ is a (non necessarily linear) function.

The interaction Hamiltonian H_I is defined as follows

$$H_I = \int \overline{g(k)} a(k) D^* dk + \text{ h.c.} \tag{4}$$

We investigate the dynamics of this system in the stochastic limit, cf. [4], in the regime of weak coupling ($\lambda \to 0$) and large times. This regime is given by time rescaling to $t \mapsto \frac{t}{\lambda^2}$. This rescaling and the interaction (4) lead naturally to introduce the rescaled quantum fields

$$\frac{1}{\lambda} e^{-\frac{it}{\lambda^2}(\omega(k)-\omega)} a(k) \tag{5}$$

where ω are the Bohr frequencies (differencies of eigenvalues of the system Hamiltonian H_S).

By the stochastic golden rule [4] the rescaled field (5) in the stochastic limit becomes a quantum white noise $b_\omega(t,k)$, or master field satisfying the commutation relations

$$[b_\omega(t,k), b^*_{\omega'}(t',k')] = \delta_{\omega,\omega'} 2\pi\delta(t-t')\delta(\omega(k)-\omega)\delta(k-k') \tag{6}$$

and with the mean zero gauge invariant Gaussian state with correlations:

$$\langle b^*_\omega(t,k) b_{\omega'}(t',k') \rangle = \delta_{\omega,\omega'} 2\pi\delta(t-t')\delta(\omega(k)-\omega)\delta(k-k') N(k) \tag{7}$$

$$\langle b_\omega(t,k) b^*_{\omega'}(t',k') \rangle = \delta_{\omega,\omega'} 2\pi\delta(t-t')\delta(\omega(k)-\omega)\delta(k-k')(N(k)+1) \tag{8}$$

White noises, corresponding to different frequencies ω, are independent.

The Schrödinger equation becomes a white noise Hamiltonian equation, cf. [4], [3] which when put in normal order is equivalent to the quantum stochastic differential equation (QSDE)

$$dU_t = (-idH(t) - Gdt)U_t \qquad ; \quad t > 0 \tag{9}$$

with initial condition $U_0 = 1$ and where

(i) $h(t)$ is the white noise Hamiltonian and $dH(t)$, called *the martingale term*, is the stochastic differential:

$$dH(t) = \int_t^{t+dt} h(s) ds = \sum_\omega \left(E^*_\omega(D)\, dB_\omega(t) + E_\omega(D)\, dB^*_\omega(t) \right) \tag{10}$$

driven by the quantum Brownian motions

$$dB_\omega(t) := \int_t^{t+dt} \int dk\overline{g}(k) b_\omega(\tau,k) d\tau =: \int_t^{t+dt} b_\omega(\tau,g)d\tau \tag{11}$$

(ii) The operator G, called the *drift*, is given by

$$G = \sum_\omega \Big((g|g)^-_\omega E^*_\omega\,(D)\, E_\omega\,(D) + \overline{(g|g)}^+_\omega E_\omega\,(D)\, E^*_\omega\,(D) \Big) \tag{12}$$

where the explicit form of the constants $(g|g)^\pm_\omega$, called the generalized susceptivities, is:

$$(g|g)^-_\omega = \int dk\,|g(k)|^2 \frac{-i(N(k)+1)}{\omega(k)-\omega-i0} = \tag{13}$$

$$= \pi \int dk\,|g(k)|^2 (N(k)+1)\delta(\omega(k)-\omega) - i\,\text{P.P.} \int dk\,|g(k)|^2 \frac{(N(k)+1)}{\omega(k)-\omega}$$

$$(g|g)^+_\omega = \int dk\,|g(k)|^2 \frac{-iN(k)}{\omega(k)-\omega-i0} = \tag{14}$$

$$= \pi \int dk\,|g(k)|^2 N(k)\delta(\omega(k)-\omega) - i\,\text{P.P.} \int dk\,|g(k)|^2 \frac{N(k)}{\omega(k)-\omega}$$

In the present paper we consider a generic quantum system, for which for each Bohr frequency ω there exist a unique pair of eigenstates $|1_\omega\rangle$ and $|2_\omega\rangle$ corresponding to the two energy levels, $\varepsilon_{1_\omega}, \varepsilon_{2_\omega}$, so that

$$\omega = \varepsilon_{2_\omega} - \varepsilon_{1_\omega}$$

In this case

$$E_\omega(D) = \langle 1_\omega|D|2_\omega\rangle |1_\omega\rangle\langle 2_\omega| \tag{15}$$

$$E_\omega(D)E^*_\omega(D) = |\langle 1_\omega|D|2_\omega\rangle|^2 |1_\omega\rangle\langle 1_\omega|$$

$$E^*_\omega(D)E_\omega(D) = |\langle 1_\omega|D|2_\omega\rangle|^2 |2_\omega\rangle\langle 2_\omega|$$

We consider a dispersion $\omega(k)$ which is ≥ 0 and, moreover, we suppose that the Lebesgue measure of the set $\{k : \omega(k) = 0\}$ equals to zero. This implies that the real part of generalized susceptivities Re $(g|g)^\pm_\omega$ is non–negative and can be non–zero only for $\omega > 0$.

We will also use the notation $(g|g)^\pm_{ij}$ for $(g|g)^\pm_\omega$ if $\omega = \varepsilon_i - \varepsilon_j$.

In the present paper we investigate the non–eqilibrium stationary states for the master equation satisfied by the diagonal part of the density matrix of a generic quantum system. This equation was obtained in [3] and is

$$\frac{d}{dt}\rho(\sigma,t) = \sum_{\sigma':\varepsilon_{\sigma'}>\varepsilon_\sigma} (\rho(\sigma',t)2\text{Re }(g|g)^-_{\sigma'\sigma} - \rho(\sigma,t)2\text{ Re }(g|g)^+_{\sigma'\sigma})|\langle\sigma, D\sigma'\rangle|^2 +$$

$$+ \sum_{\sigma':\varepsilon_\sigma>\varepsilon_{\sigma'}} (\rho(\sigma',t)2\text{Re }(g|g)^+_{\sigma\sigma'} - \rho(\sigma,t)2\text{Re }(g|g)^-_{\sigma\sigma'})|\langle\sigma', D\sigma\rangle|^2 \tag{16}$$

where $\rho(\sigma,t) = \rho(\sigma,\sigma,t)$ and $|\sigma\rangle$ are eigenvectors of the system Hamiltonian H_S.

If the system has a finite number of energy levels, there exists a stationary state for the evolution driven by the above master equation and, if the state of the reservoir is non–equilibrium, then the stationary state does not satisfy the detailed balance condition for the master equation.

The diagonal and the off–diagonal terms of the reduced density matrix evolve separately. The off–diagonal part of the density matrix evolves independently and vanishes exponentially, cf. [3]. This corresponds to the collapse of a quantum state to a classical mixed state, described by the diagonal part of the density matrix. The diagonal part $\rho(\sigma,\sigma,t)$ may be considered as a classical distribution function and equation (16) may be considered as a kinetic equation it.

The structure of the present paper is as follows.

In section 2 we describe the stationary state for a 3–level atom interacting with radiation found in [3].

In section 3 we investigate the properties of this stationary and find that the quotient of the populations of energy levels in the considered nonequilibrium stationary state does not satisfy the Einstein emission/absorption relation. We find that in particular case the quotient of populations will satisfy a new relation that we call the Double Einstein relation.

In section 4 we use the form of this state to describe the inversion of population in our 3–level system.

In section 5 we derive a master equation for the density of photons.

In section 6 we use this equation to investigate emission and absorption of radiation by the system in the non–equilibrium stationary state, obtained in [3].

2 Stationary state for 3–level system

Consider a 3–level system with energy levels $|1\rangle$, $|2\rangle$, $|3\rangle$ with energies $\varepsilon_1<\varepsilon_2<\varepsilon_3$ and Bohr frequencies

$$\omega_1=\varepsilon_2-\varepsilon_1,\quad \omega_2=\varepsilon_3-\varepsilon_1,\quad \omega_3=\varepsilon_3-\varepsilon_2$$

In the work [3] we found that the dynamics generated by the master equation (equation for reduced density matrix of the system) describes relaxation of the system to a stationary state. In this state the off–diagonal (in the basis of egenvectors of the system Hamiltonian H_S) elements of the reduced density matrix are equal to zero (this describes the effect of decoherence of a system coupled to reservoir) and the diagonal elements of the reduced density matrix for 3–level system take the form:

$$\rho_1=|\langle 1D2\rangle|^2|\langle 1D3\rangle|^2\frac{I(\omega_1)}{1-e^{-\beta(\omega_1)}}\frac{I(\omega_2)}{1-e^{-\beta(\omega_2)}}+|\langle 1D2\rangle|^2|\langle 2D3\rangle|^2\frac{I(\omega_1)}{1-e^{-\beta(\omega_1)}}\frac{I(\omega_3)}{1-e^{-\beta(\omega_3)}}+$$
$$+|\langle 1D3\rangle|^2|\langle 2D3\rangle|^2\frac{I(\omega_2)}{1-e^{-\beta(\omega_2)}}\frac{I(\omega_3)}{e^{\beta(\omega_3)}-1}\tag{17}$$

$$\rho_2=|\langle 1D2\rangle|^2|\langle 1D3\rangle|^2\frac{I(\omega_1)}{e^{\beta(\omega_1)}-1}\frac{I(\omega_2)}{1-e^{-\beta(\omega_2)}}+|\langle 1D2\rangle|^2|\langle 2D3\rangle|^2\frac{I(\omega_1)}{e^{\beta(\omega_1)}-1}\frac{I(\omega_3)}{1-e^{-\beta(\omega_3)}}+$$
$$+|\langle 1D3\rangle|^2|\langle 2D3\rangle|^2\frac{I(\omega_2)}{e^{\beta(\omega_2)}-1}\frac{I(\omega_3)}{1-e^{-\beta(\omega_3)}}\tag{18}$$

$$\rho_3=|\langle 1D2\rangle|^2|\langle 1D3\rangle|^2\frac{I(\omega_1)}{1-e^{-\beta(\omega_1)}}\frac{I(\omega_2)}{e^{\beta(\omega_2)}-1}+|\langle 1D2\rangle|^2|\langle 2D3\rangle|^2\frac{I(\omega_1)}{e^{\beta(\omega_1)}-1}\frac{I(\omega_3)}{e^{\beta(\omega_3)}-1}+$$

$$+|\langle 1D3\rangle|^2|\langle 2D3\rangle|^2\frac{I(\omega_2)}{e^{\beta(\omega_2)}-1}\frac{I(\omega_3)}{e^{\beta(\omega_3)}-1} \tag{19}$$

where $\rho_i = \langle i|\rho|i\rangle$ and

$$I(\omega) = \int |g(k)|^2\delta(\omega(k)-\omega)dk$$

3 The Double Einstein formula

Consider the quotient

$$\frac{\text{Re }(g|g)^-_\omega}{\text{Re }(g|g)^+_\omega} = \frac{N(\omega)+1}{N(\omega)} \tag{20}$$

Recalling that $N(\omega)$ is the density of field quanta (photon, phonons,...) at frequency ω, and comparing formula (20) with the well known formula of radiation theory (the Einstein formula)

$$\frac{W_{\text{emission}}}{W_{\text{absorption}}} = \frac{\overline{n}_\omega + 1}{\overline{n}_\omega} \tag{21}$$

giving the quotient of the probability of emission and absorption of a light quantum by an atom (cf. [6], Chap. V, paragraph 17, formula (18)), we gain some physical intuition of the meaning of the generalized susceptivities. In fact the quotient (21) *... is just that which is necessary to preserve the correct thermal equilibrium of the radiation with the gas ...* ([6], p.180).

In the stochastic limit approach this statement can be proved using the master equation (16) for the diagonal part of density matrix. One can prove that, if the state of reservoir is equilibrium, then the dynamics generated by the master equation describes the relaxation to equilibrium state of the system satisfying the detailed balance condition for (16) i.e.:

$$\frac{\rho_{\sigma'}}{\rho_\sigma} = \frac{\text{Re }(g|g)^-_\omega}{\text{Re }(g|g)^+_\omega} = \frac{N(\omega)+1}{N(\omega)}, \qquad \omega = \varepsilon_\sigma - \varepsilon_{\sigma'} > 0 \tag{22}$$

For equilibrium state the quotient of populations of the two levels with energy difference ω is equal to the Einstein emission–absorption quotient for quanta with energy ω. This suggests that the quotients (20) may play a similar role for some stationary non equilibrium states.

Let us give an example of such a state for which we will get a generalization of condition (22). Consider the state (17), (18), (19). For simplicity we consider the case when the matrix element $\langle 1D2\rangle$ is negligible (direct transitions between levels 1 and 2 are prohibited). In this case

$$\frac{\rho_2}{\rho_1} = \frac{\text{Re }(g|g)^+_{\omega_2}}{\text{Re }(g|g)^-_{\omega_2}}\frac{\text{Re }(g|g)^-_{\omega_3}}{\text{Re }(g|g)^+_{\omega_3}} = \frac{N(\omega_2)}{N(\omega_2)+1}\frac{N(\omega_3)+1}{N(\omega_3)} = e^{-\beta(\omega_2)}e^{\beta(\omega_3)} \tag{23}$$

Comparing with (22) and the Einstein emission–absorption relation we will call this formula the Double Einstein formula.

Relation (23) for the considered system is natural, since direct transitions from level 2 to level 1 are prohibited ($\langle 1D2\rangle$ is negligible). In this case to jump from level 2 to level 1 the system have to make two consequent jumps: from level 2 to level 3 and then from level 3 to level 1. Therefore it is natural to represent (23) in the following form:

$$\frac{\rho_1}{\rho_2} = \frac{W_{\text{absorption}}}{W_{\text{emission}}}\bigg|_{2-3}\frac{W_{\text{emission}}}{W_{\text{absorption}}}\bigg|_{1-3} = \frac{N(\omega_2)+1}{N(\omega_2)}\frac{N(\omega_3)}{N(\omega_3)+1}$$

Note that this formula is true for a special choice of the system ($\langle 1D2\rangle = 0$). Moreover, for a Gibbs state, β is linear and (23) coincides with the Einstein relation with $\frac{N(\omega_1)+1}{N(\omega_1)}$ at the RHS.

4 Inverse population state

Let us consider the following question: for a Gibbs distribution we have $\rho_1 > \rho_2 > \rho_3$: the number of particles at an energy level decreases with increasing of energy. Can we find such a stationary state where at least one pair of levels has inversed order: the number of particles increases with increasing energy? Such kind of states are important in quantum optics (laser theory).

Let us apply the method of the previous section to construct a stationary state where $\rho_2 > \rho_1$ (population of level 2 is larger than population of level 1).

We will consider the same system considered in the previous section. In particular, we take

$$\langle 1D2\rangle = 0$$

Then from (23) we have $\rho_2 > \rho_1$ if and only if

$$e^{-\beta(\omega_2)}e^{\beta(\omega_3)} > 1$$

This inequality is equivalent to:

$$\beta(\omega_3) > \beta(\omega_1 + \omega_3) \tag{24}$$

This means that the local temperature function is non–monotonic and can decrease with an increase of energy. Let us note that the quotient ρ_2/ρ_1 in the considered approximation $\langle 1D2\rangle = 0$ (metastable level 2) does not depend on $\langle 1D3\rangle$ and $\langle 2D3\rangle$. We found that for non–monotonic temperature functions we can have the inverse population effect: population of level with higher energy is larger than population of level with lower energy. In the theory of lasers the inverse population effect sometimes is discussed as an effect of negative temperature [7]. Indeed if we suppose that the state of the field is equilibrium and therefore the local temperature function is linear $\beta(\omega) = \beta\omega$ then (24) takes the form

$$\beta\omega_1 < 0$$

for $\omega_1 > 0$.

In our approach we found the inverse population effect without introduction of any negative temperature. This effect follows from the fact that the reservoir is highly non–equilibrium and the temperature function can decrease with energy.

5 Master equation for the number operator

Consider the number operator $n(k) = a^*(k)a(k)$. This operator has constant free evolution

$$e^{itH_0}n(k)e^{-itH_0} = n(k)$$

and therefore does not change in the stochastic limit. The relation with the master field is as follows

$$[b_\omega(t,k), n(k')] = \lim_{\lambda\to 0}\frac{1}{\lambda}e^{-\frac{it}{\lambda^2}(\omega(k)-\omega)}[a(k), n(k')] =$$

$$= \lim_{\lambda \to 0} \frac{1}{\lambda} e^{-\frac{it}{\lambda^2}(\omega(k)-\omega)} a(k)\delta(k-k') = b_\omega(t,k)\delta(k-k')$$

This means that the number operator extends the quantum noise algebra.

Let us find the master equation for the number operator. Since number operator does not commute with the noises we can not directly apply the master equation from [3]. Applying the QSDE for the evolution operator we get

$$d\langle U_t^* n(k) U_t\rangle = \sum_\omega \langle U_t^* \Big(E_\omega^* (D)\, dB_\omega(t) n(k) E_\omega (D)\, dB_\omega^*(t) +$$

$$+ E_\omega (D)\, dB_\omega^*(t) n(k) E_\omega^* (D)\, dB_\omega(t) - dt n(k) 2\mathrm{Re}\, G \Big) U_t\rangle =$$

$$= \sum_\omega \langle U_t^* \Big(E_\omega^* (D)\, E_\omega (D)\, [dB_\omega(t), n(k)] dB_\omega^*(t) +$$

$$+ E_\omega (D)\, E_\omega^* (D)\, [dB_\omega^*(t), n(k)] dB_\omega(t) \Big) U_t\rangle =$$

$$= dt \sum_\omega \langle U_t^* \Big(E_\omega^* (D)\, E_\omega (D)\, |g(k)|^2 2\pi\delta(\omega(k)-\omega)(N(k)+1) -$$

$$- E_\omega (D)\, E_\omega^* (D)\, |g(k)|^2 2\pi\delta(\omega(k)-\omega) N(k) \Big) U_t\rangle$$

Denoting

$$\langle X\rangle_t = \langle U_t^* X U_t\rangle$$

we arrive to master equation

$$\frac{d}{dt}\langle n(k)\rangle_t = 2\pi \sum_\omega \delta(\omega(k)-\omega) \Big(|g(k)|^2 (N(k)+1) \langle E_\omega^*(D) E_\omega(D)\rangle_t -$$

$$- |g(k)|^2 N(k) \langle E_\omega(D) E_\omega^*(D)\rangle_t \Big) \tag{25}$$

This is a completely general master equation for the number operator $n(k)$. Therefore to find $\langle n(k)\rangle_t$ it is sufficient to determine $\langle E_\omega^*(D)E_\omega(D)\rangle_t$ and $\langle E_\omega(D)E_\omega^*(D)\rangle_t$.

Since we consider a generic system by (15) equation (25) takes the form

$$\frac{d}{dt}\langle n(k)\rangle_t = 2\pi |g(k)|^2 \sum_\omega \delta(\omega(k)-\omega) |\langle 1_\omega|D|2_\omega\rangle|^2 \left((N(k)+1)\rho_{2_\omega} - N(k)\rho_{1_\omega}\right) \tag{26}$$

6 Interaction of atom with radiation in the stationary state

For a 2–level atom there is only one term in the ω–summation in (26) and the stationary state is given by (up to normalization)

$$\rho_1 = \mathrm{Re}\,(g|g)_\omega^-, \qquad \rho_2 = \mathrm{Re}\,(g|g)_\omega^+$$

and therefore the quotient satisfies the Einstein relation:

$$\frac{\rho_1}{\rho_2} = \frac{N(\omega)+1}{N(\omega)} = e^{\beta(\omega)} \tag{27}$$

If $N(k) = N(\omega(k))$ for stationary state for two–level system (26) takes the form

$$\frac{d}{dt}\langle n(k)\rangle_t = 2\pi\delta(\omega(k)-\omega)|g(k)|^2|\langle 1_\omega|D|2_\omega\rangle|^2\Big(N(\omega)(N(\omega)+1)-(N(\omega)+1)N(\omega)\Big) = 0 \tag{28}$$

that means that in the stationary state the atom is in equilibrium with the field. Vanishing of (28) is equivalent to the Einstein relation. Let us note that we did not assume that the state of the field is equilibrium.

For a 3–level system the master equation for the number of photons (25) takes the form

$$\frac{d}{dt}\langle n(k)\rangle_t = 2\pi|g(k)|^2\Big(\delta(\omega(k)-\omega_1)|\langle 1|D|2\rangle|^2\left((N(\omega_1)+1)\rho_2 - N(\omega_1)\rho_1\right)+$$
$$+\delta(\omega(k)-\omega_2)|\langle 1|D|3\rangle|^2\left((N(\omega_2)+1)\rho_3 - N(\omega_2)\rho_1\right)+$$
$$+\delta(\omega(k)-\omega_3)|\langle 2|D|3\rangle|^2\left((N(\omega_3)+1)\rho_3 - N(\omega_3)\rho_2\right)\Big) \tag{29}$$

This equation describes the balance of radiation with the 3–level atom. Consider now the stationary state of the atom. When the stationary state is equilibrium then in the RHS of (29) each term vanishes separately. This implies that in an equilibrium state the system is in detailed equilibrium with the radiation: emission equals to absorption for every frequency.

When the stationary state is non–equilibrium then the Einstein relation for populations of the levels will be not satisfied. This implies that each term in (29) will not vanish and the atom will emit and absorb radiation.

In this case (17), (18), (19) imply

$$|\langle 1|D|2\rangle|^2((N(\omega_1)+1)\rho_2 - N(\omega_1)\rho_1) =$$
$$= |\langle 1|D|2\rangle|^2|\langle 1D3\rangle|^2|\langle 2D3\rangle|^2 I(\omega_2)I(\omega_3)\frac{e^{\beta(\omega_1)-\beta(\omega_2)+\beta(\omega_3)}-1}{(e^{\beta(\omega_1)}-1)(1-e^{-\beta(\omega_2)})(e^{\beta(\omega_3)}-1)} \tag{30}$$

$$|\langle 1|D|3\rangle|^2\left((N(\omega_2)+1)\rho_3 - N(\omega_2)\rho_1\right) =$$
$$= |\langle 1|D|3\rangle|^2|\langle 1D2\rangle|^2|\langle 2D3\rangle|^2 I(\omega_1)I(\omega_3)\frac{1-e^{-\beta(\omega_2)+\beta(\omega_1)+\beta(\omega_3)}}{(1-e^{-\beta(\omega_2)})(e^{\beta(\omega_1)}-1)(e^{\beta(\omega_3)}-1)} \tag{31}$$

$$|\langle 2|D|3\rangle|^2\left((N(\omega_3)+1)\rho_3 - N(\omega_3)\rho_2\right) =$$
$$= |\langle 2|D|3\rangle|^2|\langle 1D2\rangle|^2|\langle 1D3\rangle|^2 I(\omega_1)I(\omega_2)\frac{e^{\beta(\omega_3)+\beta(\omega_1)-\beta(\omega_2)}-1}{(e^{\beta(\omega_3)}-1)(e^{\beta(\omega_1)}-1)(1-e^{-\beta(\omega_2)})} \tag{32}$$

Note that (30), (31), (32) contain the combination $\beta(\omega_3)+\beta(\omega_1)-\beta(\omega_2)$ that vanishes for an equilibrium state of the reservoir.

Equation (29) takes the form

$$\frac{d}{dt}\langle n(k)\rangle_t = 2\pi|g(k)|^2|\langle 1|D|2\rangle|^2|\langle 1D3\rangle|^2|\langle 2D3\rangle|^2\frac{e^{\beta(\omega_1)-\beta(\omega_2)+\beta(\omega_3)}-1}{(e^{\beta(\omega_1)}-1)(1-e^{-\beta(\omega_2)})(e^{\beta(\omega_3)}-1)}$$

$$\Big(I(\omega_2)I(\omega_3)\delta(\omega(k)-\omega_1) - I(\omega_1)I(\omega_3)\delta(\omega(k)-\omega_2) + I(\omega_1)I(\omega_2)\delta(\omega(k)-\omega_3)\Big) \tag{33}$$

Equation (33) shows that the 3–level system in non–equilibrium stationary state converts radiation with the energy $\omega_2 = \omega_1 + \omega_3$ into radiation with energies ω_1 and ω_3 if

$$\beta(\omega_1) + \beta(\omega_3) > \beta(\omega_1 + \omega_3)$$

and vice versa in the case of the opposite inequality.

Integrating (33) over k we get

$$\frac{d}{dt}\int E(\omega(k))\langle n(k)\rangle_t dk = 2\pi|\langle 1|D|2\rangle|^2|\langle 1D3\rangle|^2|\langle 2D3\rangle|^2$$

$$I(\omega_1)I(\omega_2)I(\omega_3)\left(E(\omega_1) - E(\omega_2) + E(\omega_3)\right)\frac{e^{\beta(\omega_1)-\beta(\omega_2)+\beta(\omega_3)} - 1}{(e^{\beta(\omega_1)}-1)(1-e^{-\beta(\omega_2)})(e^{\beta(\omega_3)}-1)} \tag{34}$$

When $E(\omega)$ is the dispersion of the field

$$E(\omega) = \omega$$

then

$$E(\omega_1) - E(\omega_2) + E(\omega_3) = 0$$

and (34) implies the conservation of energy

$$\frac{d}{dt}\int \omega(k)\langle n(k)\rangle_t dk = 0$$

For the time derivative of number operator we get

$$\int \frac{d}{dt}\langle n(k)\rangle_t dk = 2\pi|\langle 1|D|2\rangle|^2|\langle 1D3\rangle|^2|\langle 2D3\rangle|^2$$

$$I(\omega_1)I(\omega_2)I(\omega_3)\frac{e^{\beta(\omega_1)-\beta(\omega_2)+\beta(\omega_3)} - 1}{(e^{\beta(\omega_1)}-1)(1-e^{-\beta(\omega_2)})(e^{\beta(\omega_3)}-1)}$$

Using that $\omega_2 = \omega_1 + \omega_3$ we obtain that if

$$\beta(\omega_1) + \beta(\omega_3) < \beta(\omega_1 + \omega_3) \tag{35}$$

then the derivative is negative and the system absorbs the radiation (total number of absorbed photons is larger than the total number of emitted photons). In this case (33) implies that the system absorbs photons with frequencies ω_1 and ω_3 and emits photons with frequency ω_2.

If

$$\beta(\omega_1) + \beta(\omega_3) > \beta(\omega_1 + \omega_3) \tag{36}$$

then the derivative is positive and the system emits the radiation (the total number of absorbed photons is smaller than the total number of emitted photons). In this case (33) implies that the system emits photons with frequencies ω_1 and ω_3 and absorbs photons with frequency ω_2. For instance in the case of inverse population (24) the condition of emission regime (36) is satisfied.

These regimes of emission and absorption are controlled only by the difference $\beta(\omega_1)+\beta(\omega_3)-\beta(\omega_1+\omega_3)$.

Acknowledgements

The authors are grateful to I.V.Volovich for discussions. Kentaro Imafuku and Sergei Kozyrev are grateful to Centro Vito Volterra and Luigi Accardi for kind hospitality. This work was partially supported by INTAS 9900545 grant. Kentaro Imafuku is supported by an overseas research fellowship of Japan Science and Technology Corporation. Sergei Kozyrev was partially supported by RFFI 990100866 grant.

References

[1] L.Accardi, K.Imafuku, S.V.Kozyrev, *Stimulated emission with a non–equilibrium state of radiation*, Preprint Centro Vito Volterra

[2] L.Accardi, S.V.Kozyrev, I.V.Volovich, *Dynamics of dissipative two–state systems in the stochastic approximation*, Phys. Rev. A 57 N. 3 (1997); quant-ph/9706021

[3] L.Accardi, S.V.Kozyrev, Quantum interacting particle systems, Lecture Notes of Levico school, September 2000, Preprint Centro Vito Volterra N431

[4] L.Accardi, Y.G.Lu, I.V.Volovich, *Quantum theory and its stochastic limit*, Oxford Univ.Press, (in press)

[5] P. Glansdorf, I.Prigogine, *Thermodynamic theory of structure, stability and fluctuations*, Whiley Interscience, London 1971

[6] W.Heitler, *The quantum theory of radiation*, Oxford University Press, London, 1954

[7] H.Haken, In *Encyclopedia of Physics*, Vol.XXV/2c, Laser theory, Springer, Berlin–Heidelberg–New York 1970